# **REMARKS**

Claims 14-29 remain in the application. A total of 16 claims remain in the case. Independent claim 14 is amended to recite that "...the second reaction is performed under different conditions compared to that of the first reaction". Support for this new recitation in claim 14 appears in paragraph [0042] of the specification. This claim 14 is believed to be patentable for the reasons stated hereinbelow. The balance of the claims, namely claims 15-29, depends either directly or indirectly from independent claim 14 and are likewise believed to be allowable due to their dependency on an allowable base claim.

# The Claimed Invention

As recited in claim 14, the present invention is directed to a method of producing soft paper comprising adding a paper additive composition to pulp, the paper additive composition comprising an amide compound (a) or a salt thereof for softening a paper, wherein the amide compound (a) is obtained by reacting a polyamine and a carboxylic acid, the polyamine is shown by formula (1):

$$R-(NH-R^1)n-NH_2 \qquad (1)$$

wherein R is  $H_2N-R^1$  or  $R^2$  and each  $R^1$  is independently an alkylene group having 1 to 4 carbon atoms,  $R^2$  is an alkyl group or alkenyl group having 12 to 22 carbon atoms, and n is an integer of 1 to 3; the number of carbon atoms of the carboxylic acid is 10 to 24. The amide compound is obtained by reacting the carboxylic acid at a ratio of 0.5 to 4.3 moles per 1 mole of the polyamine, wherein the reaction is performed by a first reaction by reacting the polyamine and the carboxylic acid until the acid value of this reaction mixture reaches about 10% of the theoretical acid value of the initial mixture of the polyamine and the carboxylic acid and by a second reaction by allowing the reaction product of the first reaction to proceed until the acid value of this reaction mixture is further reduced to 75% or less of the acid value of the reaction product of the first reaction, and the ratio of a tertiary amine value to a total amine value of the

Application No. 10/807,092

In Response to Office Action dated April 5, 2007

Paper dated August 5, 2007

Attorney Docket No. 3274-040239

amide compound (a) is 0.60 to 0.99. This claim 14 further recites that the second reaction is performed under different conditions compared to that of the first reaction. This recitation finds support in paragraph [0042] of the specification, particularly, in the third to the last sentence in this paragraph states: "For example, a method of performing the reaction under reduced pressure after an amide compound is produced, or a method of performing the reaction at a higher temperature can be employed." Additionally, Examples 1.1 through 1.17 under the Preparation of Amide Compound, paragraphs [0083] through [0092] of the specification illustrates the second reaction being prepared under a reduced pressure.

As discussed in a previous response to an Office Action, one aspect of the present invention as recited in claim 14 is the use of a specific amide compound (a) for producing soft paper. This specific amide compound (a) of the claimed invention is obtained by reacting a specific polyamine and a specific carboxylic acid in a two-step reaction to obtain a ratio of a tertiary amine value to a total amine value ranging from 0.60 to 0.99.

## Calculation of Amine Value

Amine value represents a relative quantity of amine moieties in compounds that are contained in a sample. Amine is a compound that is obtained by substituting at least one hydrogen atom of ammonia for a hydrocarbon residue R. Amine is classified into a primary amine (i.e., RNH<sub>2</sub>), a secondary amine (i.e., RR'NH) and a tertiary amine (i.e., RR'R"N) according to the number of the hydrocarbon residue. The amine value is also classified into a primary amine value, a secondary amine value, a tertiary amine value and a total amine value that is a sum thereof.

As seen in Attachments 1 and 2, an amine value is measured by titrating with hydrochloric acid. Applicants refer the Examiner to the description on page 1, lines 1 to 4 in the right-hand column of Attachment 1, and to the description on page 1, line 8 to the bottom line in the right-hand column of Attachment 2. In this way, the amine value is measured by acids due to the basic nature of amine moiety in the compound.

On the other hand, as an amine-like compound, there are amide and imine. Amide is obtained by substituting at least a hydrogen atom of ammonia or amine for an acyl group. Imine has a C=N-R structure. Therefore, even if amide moieties or imine moieties are

Application No. 10/807,092

In Response to Office Action dated April 5, 2007

Paper dated August 5, 2007

Attorney Docket No. 3274-040239

present in a compound, the compound does not have the amine value as measured in Attachments 1 and 2.

# **Two-Step Reaction Process**

The two-step reaction of the claimed invention will be discussed with reference to Attachment 3. Attachment 3 shows a synthesis of an amide compound (W) that has tertiary amine moiety (iv) in the case of using stearic acid (X) and diethylenetriamine (Y) as raw materials. As seen in Attachment 3, the amide compound (W) that has tertiary amine moiety (iv) can be obtained by a first reaction and a second reaction. Please note that moiety (V) of the amide compound (W) is imine, but not amine.

The mixture of the two raw materials (X) and (Y) has a tertiary amine value of 0. Furthermore, the mixture has a total amine value of 250.8, a secondary amine value of 83.6 and a primary amine value of 167.2. The total amine value corresponds to the sum of two primary amine moieties (i) at both ends of diethylenetriamine (Y) and the one secondary amine moiety (ii) at the middle portion of diethylenetriamine (Y). The primary amine value corresponds to the two primary amine moieties (i) at both ends of diethylenetriamine (Y), which is obtained by subtracting the secondary amine value of 83.6 from the total amine value of 250.8.

In the first reaction, the amide compound that has two amide moieties (iii) is obtained by reacting 2 moles of stearic acid (X) with 1 mole of the diethylenetriamine (Y). In Attachment 3, an amide compound (Z) that has two amide moieties (iii) is shown as one of the main compounds obtained by the first reaction. This amide compound (Z) would be obtained when stearic acids (X) are bound to one of the primary amine moieties (i) and the secondary amine moiety (ii) of the diethylenetriamine (Y). Therefore, this amide compound (Z) has only one primary amine moiety (i) as amine moiety. On the other hand, the other main amide compound (Z'), which is not shown in Attachment 3, would be obtained when stearic acids (X) are bound to the two primary amine moieties (i) at both ends of the diethylenetriamine (Y). Therefore, this amide compound (Z') has only one secondary amine moiety (ii) as amine moiety. Since both of the amide compounds (Z) and (Z') do not have a tertiary amine moiety, each of the tertiary amine values of these amide compounds (Z) and (Z') is 0.

The amide compounds (Z) and (Z') have a total amine value of 88.3. This total amine value corresponds to the sum of the primary amine moiety (i) of the amide compound (Z) and the secondary amine moiety (ii) of the amide compound (Z'). In this way, the amide compound that has a tertiary amine moiety cannot be obtained in the first reaction, and therefore, the tertiary amine value of the amide compound obtained by the first reaction is 0.

Next, in the second reaction that followed by the first reaction, the amide compound (Z) is converted to an amide compound (W) that has a tertiary amine moiety (iv). However, as seen in Attachment 3, the second reaction is reversible while the first reaction is irreversible. That is, the amide moiety (iii) that is formed in the amide compound (Z) obtained by the first reaction is not hydrolyzed by water. However, the ring structure that is formed in the amide compound (W) obtained by the second reaction can be easily hydrolyzed by water that is produced in the second reaction. Therefore, the chemical equilibrium in the second reaction lies so far to the amide compound (Z) that has no tertiary amine moiety.

In the conventional amidation process, the second reaction has occurred however since the chemical equilibrium in the second reaction lies so far to the amide compound (Z), the amide compound (Z) is obtained as the main amide compound. Therefore, in the conventional amidation process, it is difficult to obtain the amide compound (W) that has a tertiary amine moiety (iv). As a result, the amide compound that has the ratio of a tertiary amine value to a total amine value of 0.60 to 0.99 cannot be obtained by the conventional amidation method.

In the claimed invention, in order to lay the chemical equilibrium in the second reaction so far to the amide compound (W), the second reaction is performed under the condition in which H<sub>2</sub>O is removed from the reaction system. That is, for the first reaction, a specific carboxylic acid (X) and a specific polyamine (Y) are reacted until the acid value of the reaction mixture reaches about 10% of the theoretical acid value of the initial mixture of the raw materials. Next, the second reaction is performed under different operating conditions compared to that of the first reaction, such as a reduced pressure or higher temperature compared to that of the first reaction, so as to produce the amide compound (W) in which the acid value is reached to 75% or less of the acid value of the amide compound (Z) obtained by the first reaction. By the second reaction of the claimed invention, the chemical equilibrium in the second reaction lies so far to the amide compound (W), such that the amide compound (W) that has tertiary amine

Application No. 10/807,092 In Response to Office Action dated April 5, 2007 Paper dated August 5, 2007

Attorney Docket No. 3274-040239

moiety (iv) is easily obtained. Each of a tertiary amine value and a total amine value of the amide compound (W) is 90.9, respectively, and therefore, the ratio of a tertiary amine value to a total amine value of the amide compound (W) is 1.0.

The amide compound (a) that is used in the method of the claimed invention contains a large amount of amide compound that has tertiary amine moiety, such as amide compound (W), and therefore, satisfies the ratio of a tertiary amine value to a total amine value of 0.60 to 0.99. Please refer to Examples 1.1-1.17 in Table 1 of the present specification. In the present invention such an amide compound (A) that has the ratio of a tertiary amine value and a total amine value of 0.60 to 0.99 is used for producing soft paper.

# Rejections under 35 U.S.C. §§102(b) and 103(a)

Claims 14-17 stand rejected under 35 U.S.C. §102(b) as being anticipated by or, in the alternative, under 35 U.S.C. §103(a) as being obvious over Howland et al (WO 01/59213 A1).

The Examiner's position is that Howland et al. disclose a papermaking additive composition and a method for making paper using the additive. The composition comprises an amide compound obtained by reacting one or more fatty acids and one or more polyamines according to the following formula (2):

$$H_2N-(R_1NH)_n-H (2)$$

wherein  $R_1$  is  $C_2$  to  $C_4$  alkylene and n is 2, 3, 4 or 5. Howland et al. further discloses the preferred product as having the following formula:

$$R_3NH-(R_1NR_4)_n-R_5$$
 (3)

wherein  $R_2$  is a hydrocarbon side chain of a saturated or unsaturated fatty acid;  $R_3$ ,  $R_4$  and  $R_5$  are independently selected at each occurrence from H and  $R_2C(O)$ -; and n is 2, 3, 4 or 5, provided that at least one of  $R_3$ ,  $R_4$  and  $R_5$  are H and at least one of  $R_3$ ,  $R_4$  and  $R_5$  are  $R_2C(O)$ -.

Applicants' position is that Howland et al. fail to disclose or suggest a two-step reaction process that is performed by using the specific polyamine and the specific carboxylic

acid at a specific ratio amount to produce an amide compound (a) having a ratio of the tertiary amine value to a total amine value ranging from 0.60 to 0.99, as claimed in claim 14. Furthermore, the method of Howland et al. merely discloses a conventional amidation reaction, which may correspond to only the first reaction of Attachment 3. In Howland et al., the reaction is stopped at the time the amide compound of the above formula (3) is obtained. The amide compound of the above formula (3) corresponds to the amide compound (Z) of Attachment 3 representing the claimed invention. Two N atoms in the amide compound of formula (3) can be either primary amine moiety or amide moiety, since  $R_3$ ,  $R_4$  and  $R_5$  are independently selected at each occurrence from H and  $R_2C(O)$ -. Therefore, the amide compound of Howland et al. has no tertiary amine moiety, and the tertiary amine value of Howland et al. is 0.

In this way, <u>Howland et al. disclose only the first reaction of Attachment 3</u>. With reference to Comparative Examples 1.4 to 1.9 in Table 1 of the present specification, the obtained amide compound of this first reaction product has a ratio of a tertiary amine value to a total amine value ranging between 0 and 0.4. Therefore, in the method of Howland et al, an amide compound with a ratio of a tertiary amine value to a total amine value of 0.60 to 0.99 cannot be obtained. Thus, the amide compound used in the method of producing a soft paper of the claimed invention is quite different from the amide compound disclosed in Howland et al.

The Examiner further takes the position that when n is 2, R<sub>3</sub> is H, and R<sub>4</sub> and R<sub>5</sub> are R<sub>2</sub>C(O)- in the amide compound of the above formula (3) of Howland et al. so that the ratio of tertiary amine value to total amine value is 0.67 and the ratio of acid reactant to polyamine is 3:1.

That amide compound is shown by the following formula (4):

$$\begin{array}{c|c}
R_2 & O \\
C = O & \parallel \\
N - R_1 N - R_1 N & CR_2
\end{array}$$

$$\begin{array}{c|c}
CR_2 & CR$$

Application No. 10/807,092

In Response to Office Action dated April 5, 2007

Paper dated August 5, 2007

Attorney Docket No. 3274-040239

However, when 1 mole of the above polyamine (diethylenetriamine) and 3 moles of carboxylic acid are reacted, generally, three acyl groups are bound to three N atoms of the diethylenetriamine, respectively. It is difficult to obtain an amide compound of formula (4) that. has no acyl groups on the left-hand side of the compound and two acyl groups on the right-hand side of the compound of the above formula (4) is obtained, the N atom on the left-hand side of the obtained amide compound is a primary amine moiety, and the N atom on the right-hand side and the N atom in the middle portion of the obtained amide compound are amide moieties. That is, the amide compound of formula (4) has no tertiary amine moieties, and therefore, the total amine value and the ratio of a tertiary amine value to a total amine value are 0. Therefore, the amide compound of formula (4) does not satisfy the recitation of the ratio of a tertiary amine value to a total amine value ranging from 0.60 to 0.99 as recited in claim 14 of the claimed invention.

In this way, Howland et al. fail to disclose or suggest the invention. The amide compound (a) used in the method of the claimed invention is quite different from the amide compound disclosed in Howland et al. Therefore, the method of the claimed invention is distinct from that of Howland et al. via the new recitation in claim 14 and the different characteristics, effects and/or results of the claimed invention compared to Howland et al. Thus, the claimed invention is not obvious in view of or anticipated by Howland et al.

Claims 14-29 stand rejected under 35 U.S.C. §103(a) as being obvious over Vinson et al. (US 6,162,329) in view of Dwiggins et al. (US 6,033,523) and further in view of Asakura et al. (JP 2002-275786 A).

Vinson et al. and Dwiggins et al. disclose that quaternary ammonium is used as a softening agent for paper. However, Vinson et al. and Dwiggins et al. fail to disclose or suggest the use of the amide compound as recited in claim 14 of the present invention.

Asakura et al. disclose an amide compound as an additive for making paper using recycled paper that improves the bulkiness and oil absorption of the paper. This amide compound is made from the reaction of fatty acids having from 10 to 24 carbon atoms and a polyamine compound. However, Asakura et al. also fail to disclose or suggest that a two-step reaction is performed by using the specific polyamine and the specific carboxylic acid at a specific amount ratio, by a two-step reaction, to produce the amide compound (a) having a ratio

of a tertiary amine value to a total amine value of 0.60 to 0.99 and that the amide compound (a) is used to produce soft paper as recited in claim 14.

The Examiner takes the position that in the polyamine compound disclosed in Asakura et al., when n is 1, and the middle amine and the two hydrogen atoms on one end of the amine are substituted with acyl groups, the ratio of the tertiary amine to the total amine is 0.66. That compound is represented by formula (5):

$$\begin{array}{c|c}
R_{3} & O \\
\hline
R_{3} & C = O \\
\hline
N - R^{2}N - R_{1}N & CR \\
\hline
CR & CR
\end{array}$$
(5)

However, it is difficult to obtain the amide compound (a) of the claimed invention from the above formula (5) for the same reasons discussed hereinabove regarding formula (4) of Howland et al. Furthermore, even if the amide compound of formula (5) is obtained, the amide compound of formula (5) has no tertiary amine moiety, and therefore the total amine value and the ratio of a tertiary amine value to a total amine value are 0. Therefore, the amide compound of formula (5) does not satisfy the ratio of a tertiary amine value to a total amine value ranging from 0.60 to 0.99 as recited in claim 14 of the claimed invention.

Therefore, Applicants submit that Vinson et al., Dwiggins et al., and Asakura et al. fail to disclose or suggest the use of the amide compound (a) as recited in amended claim 14 of the claimed invention. Thus, the claimed invention particularly as recited in claim 14 is not obvious in view of the references to Vinson et al., Dwiggins et al., and Asakura et al., when considered alone or in combination.

Claims 15-29 depend either directly or indirectly from and add further limitations to claim 14. These claims are believed to be in condition for allowance since they depend from claim 14 which is believed to be in condition for allowance.

# Conclusion

In view of the above amendments and remarks, reconsideration of the rejections and allowance of claims 14-29 are respectfully requested.

Respectfully submitted,

THE WEBB LAW FIRM

**з**у\_\_\_

Kent E. Baldauf, Jr.
Registration No. 36,082
Attorney for Applicants
700 Koppers Building
436 Seventh Avenue

Pittsburgh, Pennsylvania 15219

Telephone: 412-471-8815 Facsimile: 412-471-4094

E-mail: webblaw@webblaw.com

Attachments 1-3

# **AOCS Official Method Tf 1b-64**

Replaces Nb 4-62 Reapproved 1993

# Total Amine Value of Fatty Amines, Indicator Method

Definition: The total amine value is the mg of potassium hydroxide equivalent to the basicity in 1 g of sample.

Scope: Applicable to fatty amines.

## **Apparatus**

1. Erlenmeyer flask—300 mL.

#### Reagents

- 1. Isopropyl alcohol—reagent grade.
- Bromphenol blue indicator solution—0.2% in isopropyl alcohol.
- Hydrochloric acid—approximately 0.2 N, in isopropyl alcohol, accurately standardized with sodium carbonate, using 0.1% bromcresol green as the indicator, or standardized according to AOCS Specification H 14-52.

#### Procedure

- 1. Melt the sample if it is not already liquid, mix thoroughly and weigh 0.5 g into 250-mL beaker.
- Add 50 mL of isopropyl alcohol, and boil for 1 minute to drive off any free ammonia that may be present. Cool to room temperature.

3. Add 5-10 drops of indicator, and while swirling the flask, titrate with 0.2 N HCl to the yellow end point.

## Calculation

Total amine value =  $\frac{mL \times N \times 56.1}{\text{sample wt, g}}$ 

#### Precision

At a confidence level of 95%, in the range of values of:	Amines 75-267	
1. Two single determinations performed in	-	
I lab shall not differ from the true mean		
value by more than:	± 2.8%	
2. Single determinations performed in two		
different labs shall not differ from the true		
mean value by more than:	± 3.1%	

# **AOCS Official Method Tf 2b-64**

Replaces No 4-67 Revised 1987 Responsed 1993

# Primary, Secondary, and Tertiary Amine Values of Fatty Amines, Indicator Method

Definition: The primary amine value is the mg of potassium hydroxide equivalent to the primary amine basicity in 1 g of sample. The secondary amine value is the mg of potassium hydroxide equivalent to the secondary amine basicity in 1 g of sample. The tertiary amine value is the mg of potassium hydroxide equivalent to the tertiary amine basicity in 1 g of sample.

Scope: Applicable to fatty amines. The method is not applicable to fatty amines containing reactive groups and structures, e.g., diethylene-(DETA)-diamides and 3,3'-iminobispropylamine-(DPTA)-diamides.

**Apparatus** 

- 1. Erlenmeyer flask-300 mL or beaker, 150 mL.
- 2. Magnetic stirrer—with Teflon<sup>TM</sup>-coated stirring bar.

Reagents

- Hydrochloric acid approximately 0.2 N and 0.1 N in isopropyl alcohol—accurately standardized with sodium carbonate, using 0.1% bromcresol green as the indicator, or standardized according to AOCS Specification H 14-52.
- 2. Salicylaldehyde—AR grade.
- 3. Phenyl isothiocyanate—AR grade.
- Chloroform—ACS or AR grade (see Notes, Caution and 2)
- 5. Bromphenol blue indicator solution—0.2% in 99% isopropyl alcohol.

**Procedure** 

- Determine the total amine value of AOCS Method Tf 1b-64.
- Determine the size of the sample from the following table:

Amine Value	Weight of Sample		
≤ 10	5 g		
> 10	2 g		

 Melt the sample if it is not already liquid, mix thoroughly and accurately weigh the designated size of the sample into two 250-mL beakers or 300-mL flasks. Mark them S and T.

- 4. Add 50 mL chloroform (see Notes, Caution and 2) to each flask (or beaker) and boil for 1 min on a hot plate to drive off any free ammonia that may be present.
- 5. Cool to room temperature. Add 3 mL of salicylaldehyde to sample S, and allow to stand for 5 min. Add 5 mL of phenyl isothiocyanate to sample T and allow to stand for 30 min.
- 6. Add 1 mL of indicator to each sample and while swirling the flasks (or by using the magnetic stirrer if beakers are used), titrate with 0.2 N HCl to the yellow end point (see Notes, 1). The yellow may fade back to green upon standing, but this is to be disregarded if the yellow color is bright and the addition of another mL of 0.2 N HCl does not change the yellow color.

Calculations

- Total amine value of primary, secondary and tertiary amine groups—determined by AOCS Method Tf 1b-64.
- 2. Titration S:

Amine value of secondary and tertiary amine groups =

$$mL \times N \times 56.1$$
 sample wt, g

3. Titration T:

Amine value of tertiary amine groups =

$$mL \times N \times 56.1$$
 sample wt, g

- Primary amine value = Total amine value 1 amine value 2
- 5. Secondary amine value = Amine value 2 amine value 3
- 6. Tertiary amine value = Amine value 3

#### Precision

At a confidence level of 95%, in the range of values indicated:

Types of Amine Range of Values	Primary		Secondary	Tertiary	
	0.1	5–256	8–105	0.9-1.5	68
Two single determinations performed in 1 laboratory shall not differ from the true mean value by more than:	±180%	± 2.9%	± 6.3%	± 67.5%	± 3.1%
Two single determinations performed in different laboratories shall not differ from the true mean value by more than:	± 187%	± 2.9%	± 8.0%	± 176	± 10.4%

Page 1 of 2

ATTAChMENT 2

# Primary, Secondary, and Tertiary Amine Values

Tf 2b-64

# Notes

#### Caution

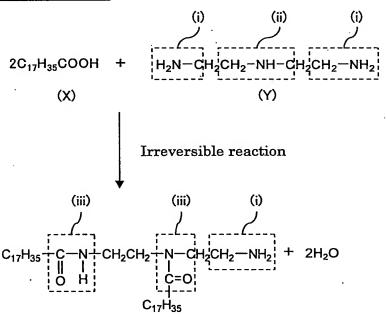
Chloroform is a known carcinogen. It is toxic by inhalation and has anesthetic properties. Avoid contact with the skin. Prolonged inhalation or ingestion can lead to liver and kidney damage and may be fatal. It is nonflammable, but will burn on prolonged exposure to flame or high temperature. The TLV is 10 ppm in air. A fume hood should be used at all times when using chloroform.

#### **Numbered Notes**

- 1. In the case of titrating the tertiary content of a primary amine it is advisable to use a microburette as the titration will be extremely small. Errors in titration are
- greatly magnified because of the high molecular weight involved. 0.1 N HCl may be used if the quantity of tertiary amine is very low.
- 2. 1,1,1-Trichloroethane (TCE) has been suggested as a replacement for chloroform in this method. Although TCE is a halogenated hydrocarbon, the TLV (350 ppm) is less that those of chloroform, carbon tetrachloride and methylene chloride. Cyclohexane and isooctane may also be considered as alternate solvents. These solvents have not been collaboratively studied within the AOCS technical committees. This recommendation does not represent official approval by the AOCS Uniform Methods Committee.

# Two-step reaction of the present invention

# (I) First reaction

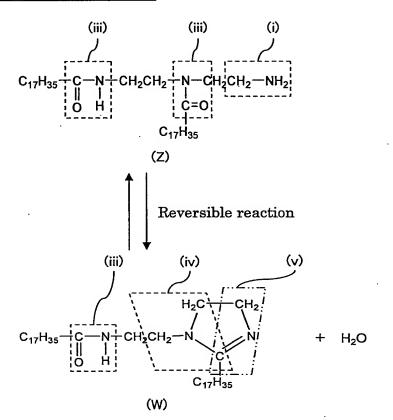


(Z)

Total amine value: 250.8
Tertiary amine value: 0
Secondary amine value: 83.6

Total amine value: 88.3 Tertiary amine value: 0 Secondary amine value: 0

# (II) Second reaction



Total amine value: 90.9 Tertiary amine value: 90.9 Secondary amine value: 0